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Published:

— with international search report

For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

(54) Title: UREA MOULDING COMPOUNDS WITH ANTISEPTIC ACTIVITY

(57) Abstract: Urea moulding compounds with antiseptic activity, the processes for the preparations thereof and the articles obtained by moulding said compounds.

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UREA MOULDING COMPOUNDS WITH ANTISEPTIC ACTIVITY

The present invention relates to urea moulding compounds with antiseptic activity, the processes for the preparation thereof and the articles obtained by moulding said compounds.

5 TECHNOLOGICAL BACKGROUND

Urea moulding compounds is traditionally obtained by preparing a urea formaldehyde resin in the presence of cellulose, then subjecting the resulting product to drying, milling and optional addition of pigments,
10 lubricants or other suitable excipients, depending on the intended applications. The urea resin is usually prepared in the presence of catalysts such as zinc salts.

The resulting product, in the form of powders or granules, can then be subjected to moulding with
15 conventional techniques, for example by transfer, injection or compression, to obtain a wide variety of articles, such as sanitary articles, kitchenware, tableware, ornamental articles, electrotechnology components and the like.

20 Sanitary articles in particular are at present one of the most important application fields for urea moulding compounds. The possibility of giving antiseptic properties to the articles obtained from urea moulding compounds would undoubtedly provide, advantages in that sanitary
25 articles having improved sanitary characteristics for the users could be manufactured. This is the case of the articles installed in public baths, for example in schools, hospitals, public premises and the like, where the need for keeping cleanness and asepsis characteristics
30 is particularly felt.

SUMMARY OF THE INVENTION

It has now been found that urea moulding compounds

can be added with suitable antiseptic agents in order to give antiseptic and biocide properties said moulding compounds as well as the articles prepared therefrom. This addition does not affect the characteristics of the moulding compounds and the biocide/antiseptic activity is surprisingly retained by the final product (mass and molded article).

Therefore, the invention in a first aspect provides urea moulding compounds based on cellulose and urea formaldehyde resin containing antiseptic agents.

The invention further relates to the articles obtainable by moulding urea compounds containing the antiseptic agents and to the processes for the preparation of the urea moulding compounds.

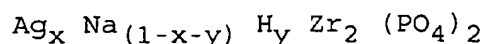
15 DETAILED DISCLOSURE OF THE INVENTION

The antiseptics which can be used according to the invention belong to the following classes:

- a) inorganic derivatives containing Ag, Cu, Zn, Fe, and the like and/or salts or complexes thereof;
- 20 b) organic derivatives containing phenol groups and halogen atoms such as Cl, Br, I;
- c) phthalate derivatives;
- d) a compound selected from N-trichloromethylthioipthalimide; 10,10'-oxybispheno-xarsine; 2-N-octyl-4-isothiazolin-3-one; N-(trichloromethylthio)-4-cyclohexene-1,2-dicarboximide; tribromosalicylamide; trichlorocarbanilide;
- 25 e) quaternary ammonium derivatives (cetrimide, dequalinium chloride).

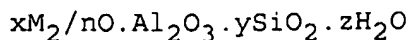
30 Examples of inorganic derivatives are the compounds:

1) Silver, Sodium, Hydrogen, Zirconium, phosphate with Ag content ranging from 3.8% to 10%, preferably compounds of formula



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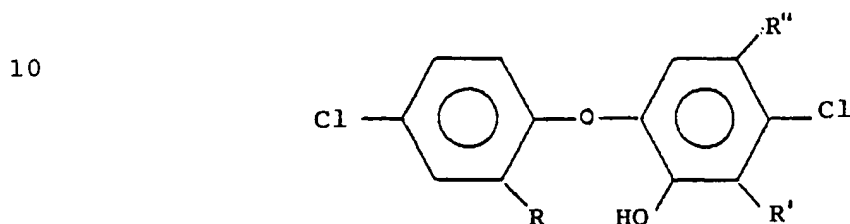
2) Aluminium silicates, preferably compounds of formula



wherein M is Ag, Na, and the like, and

5 x, y, z, are partial molar amounts.

Examples of organic derivatives containing phenol groups and halogen groups are diiodohydroxyquinoline, 2'-hydroxytrichlorodiphenyl ethers of formula



wherein

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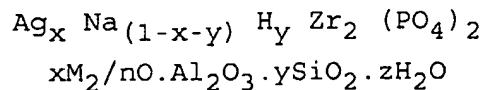
R=Cl	R'=H	R''=H
R=Cl	R'=Cl	R''=H
R=H	R'=H	R''=Cl
R=Cl	R'=Cl	R''=Cl

Examples of phthalate derivatives are:

20 Diisopropyl phthalate; dibutyl phthalate; dihexyl phthalate.

Said antiseptic agents can optionally be used in a mixture thereof.

25 Particularly preferred are inorganic antiseptics, in particular the compounds of formula



wherein M is Ag, Na, and the like, and

x, y, z, are partial molar amounts.

30 Said antiseptic agents can be present in the moulding compounds in weight percentages ranging from 0.0001 to 10%, preferably from 0.001 to 5%, more preferably from 0.01 to 1%.

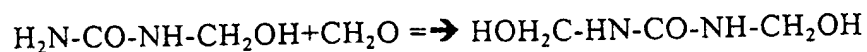
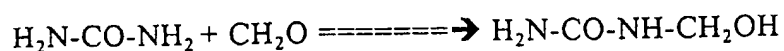
The moulding compounds of the invention can be

prepared by adding the desired amount of one of the antiseptic agents described above during any one of the operative steps of a conventional process for the preparation of urea moulding compounds.

5 The addition will preferably be carried out during the ball milling or colouring step to ensure the uniform dispersion of the antiseptic agent inside the mass. It has anyway been observed that the antiseptic agent does not adversely affect the polymerization or the cross-linking
10 of the urea formaldehyde resin and therefore the addition can also be carried out during any production step. The addition can also be carried out before the final use (moulding).

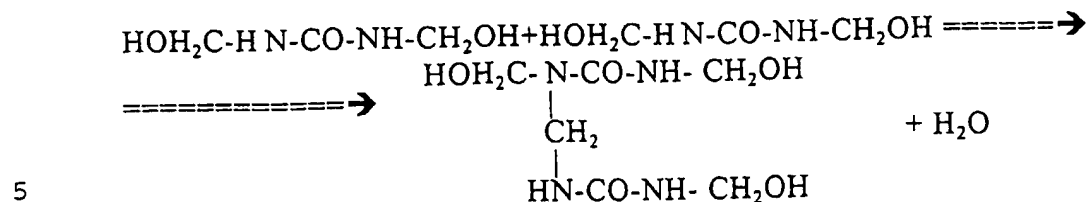
A typical procedure according to the invention for
15 the industrial production of urea moulding compounds added with antiseptic agents will now be described in greater detail.

The urea resin is prepared starting from formaldehyde and urea in a ratio of 1.00 + 3.00 moles of formaldehyde
20 per mole of urea, in aqueous medium at about neutral pH, according to the following scheme:

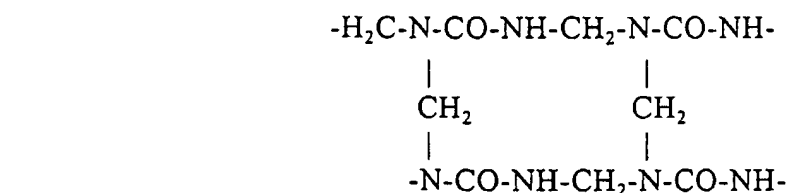


25 In this step formaldehyde is added to urea to yield methylol end groups.

Subsequently the reaction system is added with the catalyst (for example Zinc sulphate $\text{ZnSO}_4 \cdot x\text{H}_2\text{O}$ and/or Zinc sulphite $\text{ZnSO}_3 \cdot x\text{H}_2\text{O}$), which by hydrolysis yields a
30 slightly acidic environment, thus promoting the progress of the polycondensation reaction and the formation of the first cross-links with methylene bridges:



During the moulding step, cross-linking is completed by the action of high temperature and pressure, until substantially total conversion of the methylol groups into methylene bridges:



In practice, in the industrial preparation of the resin, instead of only formaldehyde, a mixture of the latter with a partially resin preformed resin is used, (which is named Formurea) in a formaldehyde/formurea 1:3 weight ratio varying depending on the intended characteristics.

The production comprises the following steps:

1) Reaction

The reagents, 36% formaldehyde solution, formurea, 70% urea solution and 32% hexamine solution, are continuously fed in fixed ratios to a maturation tank through metering pumps. The amount of hexamine solution can be varied to adjust the resin final pH from 6.0 to 8.0 (also depending on the acidity characteristics of the fed cellulose).

Afterwards, the hot urea solution is mixed with the mixture of the three other solutions in a mixing system, after that the reaction mixture is cooled down to temperatures ranging from 20° to 40°C in a water

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exchanger. The pre-condensed solution – commonly referred to as "syrup" – remains for about 1 hour under atmospheric pressure, at temperatures from 20° to 40°C.

By way of example, the percentage formulation of the syrup corresponding to a FA/U (FormAldehyde/Urea) ratio of 1.50 moles/moles.

100% Total Formaldehyde	30.02
100% Total Urea	39.62
100% Total Hexamine	1.49
10 Total water	28.87

	100.00

2) Mixing with cellulose and catalysis

The urea syrup is continuously fed to the first of a series of three mixing machines. At the same time, cellulose chips (about of 5x3x1 mm size), powdery catalyst (such as Zinc sulphate monohydrate) and a powdery pure lubricant (such as Zinc stearate) are fed in suitable amounts.

The mix may be added, in correspondence with the second mixer, with a soaking liquid, to promote the resin impregnation of cellulose.

The impregnation process is carried out under mild, protracted mixing (2 hour) at temperatures of about 40÷50°C, controlled by means of thermo-static water loop.

The formulation of a mix is reported by way of example, based on 100 kg of syrup.

Syrup	100.00 kg
93% Cellulose	25.71
30 Zinc sulphate Catalyst	0.30 (mean value)
Zinc stearate Additive	0.40
Soaking Additive	0.10

Total	126.51 kg

3) Drying

The mix is fed to a drier wherein water, both that already present in the various starting compounds and that formed during the polycondensation reaction, is removed by current of hot air (previously treated in a humidification column and then heated to temperatures ranging from 100 to 150°C in a vapour exchanger). During this step the product is subjected to high temperature for prolonged times (total permanence time about 2 hours) and the condensation reaction further proceeds. The fluidity characteristics depend on the condensation degree as well as the residual water content in the product (dry "crumb").

In the end part of the drier, the granular product of various cut shapes is cooled through a cold air flow.

The drier is operated under slightly depression compared with the outer environment.

4) Premilling and filtration

The crumb from the previous step, possibly from a storage mixer, is fed to a pulverizer mill equipped with a classifying system for particle size distribution.

Milling is carried out in air flow at controlled temperature to remove the product while preventing material from overheating.

The air exiting the mill, containing the finely powdered product (about 100 micron size) is filtered in automated cleaning bag filters, the separated product is fed to horizontal ribbon blenders (to accumulate and homogenize the semi-finished product).

The resulting product is usually referred to as "base".

100 kg of mix usually yield about 65 kg of dry base.

5) Milling, colouring, addition of the antiseptic

The base product is taken from the mixers in fixed amounts through weighing (usually 500 kg) and loaded into

ball mills with a series of successive operations: each transfer is preferably effected by pneumatic conveying equipments using compressed air.

5 The product is further milled in ball mills, while dispersing pigments (for colouring the mass) and excipients, fluidifiers, plasticizers, accelerators, antiseptics, and the like, which are added manually. Milling is carried out in batch mode for a time of about 2+6 hours).

10 The coloured powders containing the antiseptic are poured into a mixer of such a capacity as to homogenize 2 or 3 single batches, which acts as a stockage for the successive continuous operations.

6) Powder bag filling

15 According to the commercial requirements, the powdery coloured product containing the antiseptic can be packaged in bags and directly sold. Bag filling is preceded by sieving.

7) Dry-granulation

20 More frequently, the coloured powders are sieved, then compounded by the action of pressure and temperature (dry granulation). This process is carried out in screw extruders equipped with thermo-static loops, in which the powders are heated to the plastication point (80-100°C) and the mass is extruded in the form of chips, through a
25 die-cutter system.

Afterwards, chips are air cooled in vibratory units, then ground in finish mills such as toothed disc mills, cross beater mills or pil mills wherein the compacted
30 product size is further reduced. Considering the low selectivity of milling, this step should be followed by a sieving step to separate the finished product of the desired size (particle size distribution usually being 0.2 - 1.2 mm). Fines exiting the sieve are recycled to the

extruder (wherein they are combined with fresh powders),
whereas courses are directly recycled to the finish mill.

The obtained product is then homogenized in double
cone mixers wherein, if desired, the post-mixing
5 excipients, such as antiseptics, can also be dispersed.

The dry granulated product usually has bulk density
from 0.60 to 0.75 kg/l.

8) Wet granulation

Wet granulation of powders can also be carried out in
10 humid, by adding deionized water to densifiers, wherein
powders are heated to temperatures of 70 - 80°C, and
contacted with small amounts of water to form spherical
granules of very variable size (about 1 mm to 30 mm).

Granules are cooled, then fed to a disc mill and then
15 to a classifying sieve, by a series of unitary operations
quite similar to that of the dry granulation process.

The resulting product has usually bulk density values
of 0.50 kg/l to 0.60 Kg/l.

The biocide activity of the compounds of the
20 invention was assayed by the "Film contact Method" test
(Antibacterial Strength Testing Method (I) of
antibacterial processed products (1955 Edition). Film
Contact Method, SEK, Japan).

A 5x6x0.3 cm resin plate is contacted with a
25 suspension of 10^6 microorganisms/ml (1200 μ l).
Microorganisms are tested at time zero (basal) and after
24 hours.

The various antiseptics present in the resins at the
assayed concentrations (up to 10%), have surprisingly
30 shown decreases in the bacterial count from 4 to 5 log.
towards the following microbial strains:

Escherichia coli

Staphylococcus aureus

Klebsiella pneumoniae

10

Pseudomonas aeruginosaSalmonella typhimuriumProteus indole positiveStaphylococcus pyogenes5 Streptococcus faecalisCandida albicansAspergillus niger

10 The results obtained with Salmonella typhimurium, Klebsiella pneumoniae and Staphylococcus aureus are shown in the enclosed Figures 1, 2 and 3.

The addition of particular antiseptics allows therefore to obtain moulding compounds having remarkable biocide activity against a wide spectrum of pathogenic microbial strains.

15 The method of the invention may of course be subjected to changes completely or in part, depending on parameters such as the production plant or the operative scale. For example, the preparation of the mix and of the syrup can be carried out in batch, drying can be effected
20 in belt or rotary ovens, colouring can be carried out on the humid mix or in a turbomixer, wet granulation can be effected by pelletization or the compounds can be obtained by simple mixing of powdery cellulose and powdery urea/formaldehyde resin. During the condensation reaction,
25 starting materials of different physical state than those mentioned above can be used, for example gaseous formaldehyde, solid urea, hexamine in crystals. The urea, formaldehyde, formurea and hexamine ratios can range within wide limits and are anyway conventionally
30 determined by those skilled in the art.

The following Example illustrates the invention in greater detail.

EXAMPLE

49.6 kg of formurea type F630 are intimately mixed

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with 16.6 kg of 36% formaldehyde, and 1.6 kg of 32% hexamine solution during 1h at room temperature. The resulting clear solution is in its turn mixed together with 48.8 kg of 70% urea solution for 1 h. The resulting
5 syrup is first cooled to 20°C then left to stand at said temperature for about one hour. The thus treated syrup is combined with 30 kg of α cellulose cut into small pieces, 0.3 kg of zinc stearate, and zinc sulphate (sufficient to adjust the final pH to 7.3) in 1h. The mix is continuously
10 mixed for 2h at a temperature of 45°C to obtain a homogeneous but friable mass weighing about 147 kg, which is loaded as a thin layer on a rotary drier in 1h. The hot air drying step lasts about 2h to obtain a final yield of about 100 kg of dry crumb. The resulting 100 kg of crumb
15 are milled in a pulverizer mill with cold air flow to remove the product while preventing it from overheating. Air from the mill and containing the powdery product with particle size distribution ranging from 20 μ to 120 μ is filtered through bag filters; the separated product is
20 pneumatically conveyed to a 400 l ball mill containing 250 kg of porcelain balls. In addition to the 100 kg of base, the ball mill is loaded with 0.8 kg of zinc stearate, 0.1 kg of o,p-Toluenesulfonamide, 0.8 kg of Titanium dioxide; in this step the biocide is added: 1 kg of Silver-sodium-hydrogen-zirconium phosphate. The mass is rotated at 20
25 revolutions/min. for 4h keeping the inner temperature below 60°C. At the end of the operation, 102.7 kg of white powder are obtained which is poured into a 400 l ribbon mixer before being dry granulated.

30 The powder containing the biocide is compacted in about 40 minutes through a single screw extruder thermostated at 80°C, keeping the screw at 52 revolutions/min. and the absorbed power at 36 KW/h. The extruded chip is cooled to 30°C with air in a vibratory

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unit then is ground in a grinding mill rotating at 270 revolutions/min. and sieved to the desired particle size distribution (0.2 mm to 1.2 mm) through a vibrating sieve. During sieving, in addition to the desired particle size fraction, two further fractions are obtained: one with size larger than 1.2 mm and one with size smaller than 0.2mm. The first one is remilled while the second is removed.

The final yield is 80 kg of finished, packed product.

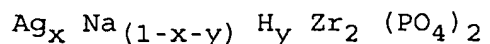
CLAIMS

1. Urea moulding compounds based on cellulose and urea/formaldehyde resin containing antiseptic agents.

2. Moulding compounds as claimed in claim 1, wherein the antiseptic agents are selected from the group consisting of: inorganic compounds containing Ag, Cu, Zn, Fe, salts or complexes thereof; organic compounds containing phenol groups and halogen atoms; phthalate derivatives; a compound selected from N-trichloromethylthiophthalimide; 10,10'-oxybisphenoxarsine; 2-N-octyl-4-isothiazolin-3-one; N-(trichloro-methylthio)-4-cyclohexene-1,2-dicarboximide; tribromosalicylamide; trichlorocarbanilide; quaternary ammonium derivatives.

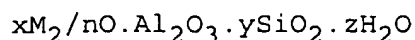
3. Moulding compounds as claimed in claim 2, wherein the antiseptic agents are selected from the group consisting of silver, zirconium and iodine hydrogen phosphates, diiodohydroxyquinoline, 2'-hydroxychlorodiphenyl ethers, diisopropyl phthalate, dibutyl phthalate, dihexyl phthalate, cetrimide, dequalinium chloride, N-trichloromethylthiophthalimide; 10,10'-oxybisphenoxarsine; 2-N-octyl-4-isothiazolin-3-one; N-(trichloro-methylthio)-4-cyclohexene-1,2-dicarboximide, tribromo-salicylamide; trichlorocarbanilide.

4. Moulding compounds as claimed in claim 1, 2, 3, wherein the antiseptic agents are compounds of formula



wherein x, y, z, are partial molar amounts.

5. Moulding compounds as claimed in claim 1, 2, 3, 4, wherein the antiseptic agents are compounds of formula



wherein M is Ag, Na, and the like, and

x, y, z, are partial molar amounts.

6. Moulding compounds as claimed in any one of claims 1

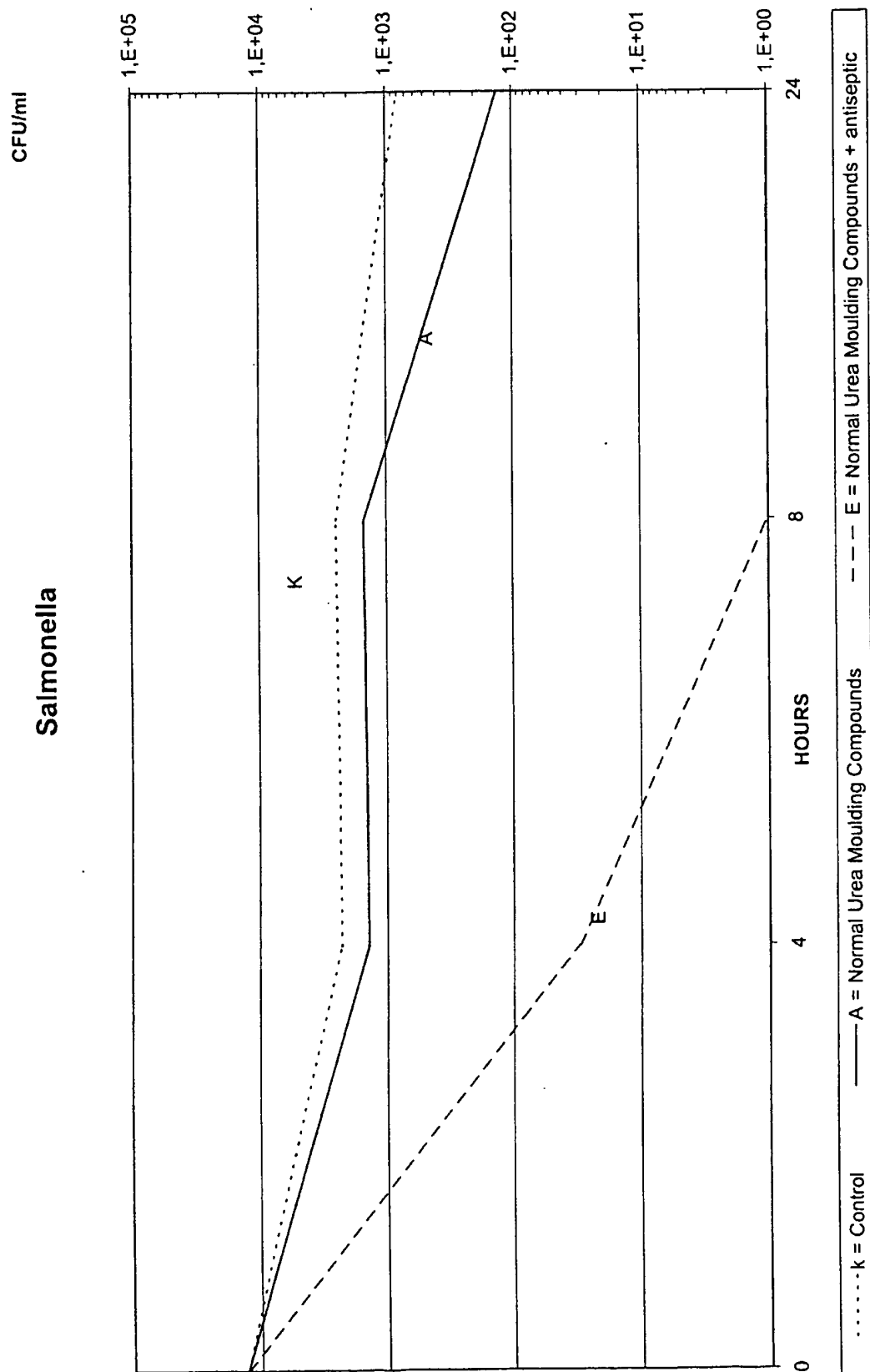
to 5, wherein the antiseptic agents are present in amounts ranging from 0.00001 to 10% by weight.

7. Articles obtainable by moulding the compounds of claims 1 to 6.

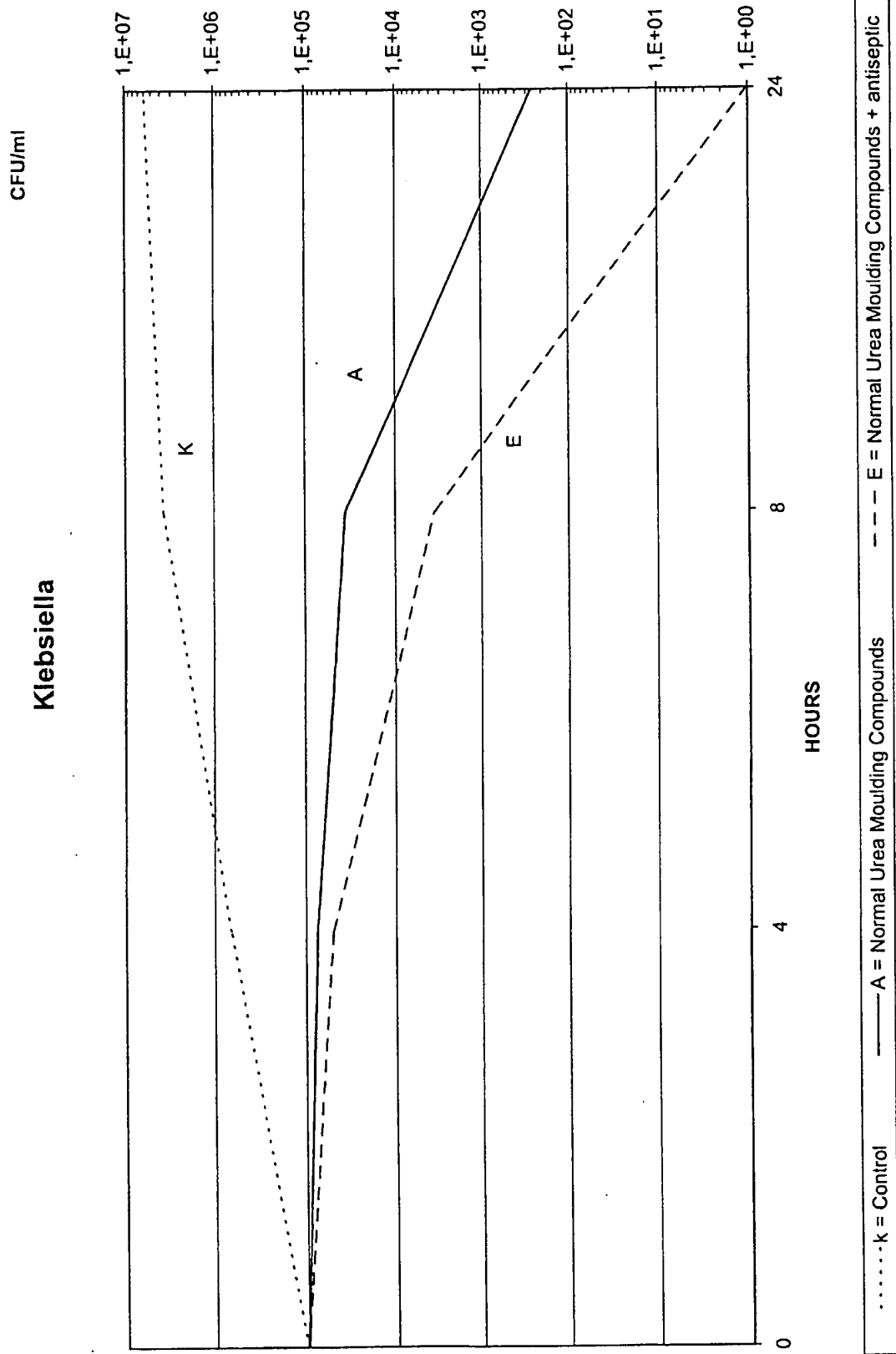
5 8. A process for the preparation of the urea moulding compounds of claims 1 to 7 comprising the addition of an antiseptic agent during one or more of the following steps:

- 10 a) preparation of a pre-condensed solution by reacting formaldehyde and urea in the presence of tertiary amines (hexamine);
 - b) mixing of the pre-condensed solution with cellulose and adding a cross-linking catalyst;
 - c) drying of the mass;
 - 15 d) premilling and filtration;
 - e) milling and optional colouring;
 - f) powder bag filling or dry- or humid- granulation;
 - g) homogenization and bag filling of the finished product.
- 20 9. A process as claimed in claim 8 wherein the addition of antiseptic agent is carried out in the milling and colouring step.
10. A process as claimed in claims 8 and 9 wherein the antiseptic agent is added before the final use (moulding).

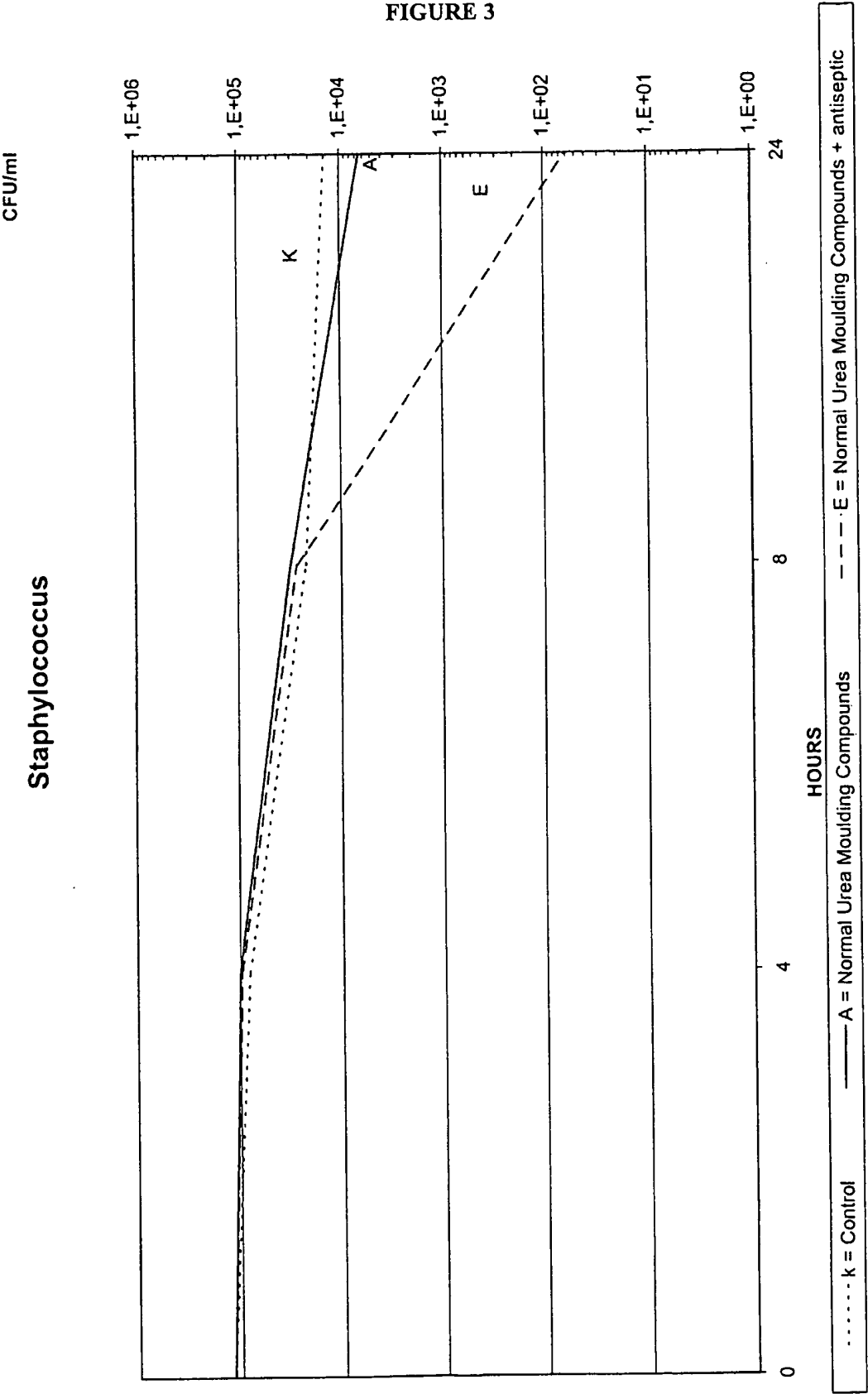
1/3
FIGURE 1



2/3
FIGURE 2



3/3
FIGURE 3



INTERNATIONAL SEARCH REPORT

International Application No

PCT/EP 00/04966

A. CLASSIFICATION OF SUBJECT MATTER

IPC 7 C08L61/24 C08G12/12 C08K3/00 C08K5/00

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 C08L C08G C08K

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal, WPI Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 5 393 809 A (GUERET JEAN-LOUIS H) 28 February 1995 (1995-02-28) claim 1	1,6,7
Y	column 3, line 54 - line 67 ---	2-5,8-10
Y	US 5 296 238 A (SUGIURA KOJI ET AL) 22 March 1994 (1994-03-22) column 3, line 32 claim 1 column 1, line 12 - line 14 ---	2-4
Y	EP 0 444 939 A (HAGIWARA RESEARCH CORP ;JAPAN ELECTRONIC MATERIALS (JP)) 4 September 1991 (1991-09-04) page 3, line 25 page 4, line 49 --- -/-	5

☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

* Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"&" document member of the same patent family

Date of the actual completion of the international search

19 December 2000

Date of mailing of the international search report

16. 03. 2001

Name and mailing address of the ISA

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Authorized officer

Paalman, R

INTERNATIONAL SEARCH REPORT

International Application No

PC, L. 00/04966

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT		
Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	<p>US 3 516 953 A (WOOD ERNEST HERBERT)</p> <p>23 June 1970 (1970-06-23)</p> <p>example 1</p> <p>-----</p>	8-10

INTERNATIONAL SEARCH REPORT

International application No.
PCT/EP 00/04966

Box I Observations where certain claims were found unsearchable (Continuation of item 1 of first sheet)

This International Search Report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:
because they relate to subject matter not required to be searched by this Authority, namely:
2. ☐ Claims Nos.:
because they relate to parts of the International Application that do not comply with the prescribed requirements to such an extent that no meaningful International Search can be carried out, specifically:
3. ☐ Claims Nos.:
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

Box II Observations where unity of invention is lacking (Continuation of item 2 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

see additional sheet

1. ☐ As all required additional search fees were timely paid by the applicant, this International Search Report covers all searchable claims.
2. ☐ As all searchable claims could be searched without effort justifying an additional fee, this Authority did not invite payment of any additional fee.
3. ☐ As only some of the required additional search fees were timely paid by the applicant, this International Search Report covers only those claims for which fees were paid, specifically claims Nos.:
4. ☒ No required additional search fees were timely paid by the applicant. Consequently, this International Search Report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

1,4,5 and 2,3,6-10 partially

Remark on Protest

- ☐ The additional search fees were accompanied by the applicant's protest.
- ☐ No protest accompanied the payment of additional search fees.

INTERNATIONAL SEARCH REPORT

International Application No. PCT/EP 00/04966

FURTHER INFORMATION CONTINUED FROM PCT/ISA/ 210

This International Searching Authority found multiple (groups of) inventions in this international application, as follows:

1. Claims: 1,4,5 and (2,3,6-10 partially)

Urea moulding compositions comprising urea-formaldehyde, cellulose and an antiseptic agent, the antiseptic agent being selected from an inorganic compound containing Ag, Cu, Zn, Fe and the like and/or salts thereof;

2. Claims: 2,3, 6-10 (in part)

wherein the antiseptic agent is an organic derivative containg phenol groups and halogen atoms;

3. Claims: 2,3, 6-10 (in part)

wherein the antiseptic agent is a phthalate derivative;

4. Claims: 2,3, 6-10 (part)

wherein the antiseptic agent is chosen among some specific defined compounds;

5. Claims: 2,3,6-10 (in part)

wherein the antiseptic agent is chosen among quaternary ammonium derivatives.

INTERNATIONAL SEARCH REPORT

Information on patent family members

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